Chapter 5

Conclusions

5.1 Aims

In this thesis I aim to improve understanding of natural contributions to ozone over Australia and the southern ocean. Chapter 1 Introduction and Literature Review examines processes leading to ozone creation in the troposphere, primarily isoprene emissions and subsequent chemistry. A summary of how the lack of information available over Australia affects modelling and forecasting is provided. The secondary cause of ozone, stratosphere to troposphere transport (STT), is also outlined with associated processes and causes discussed. Methodologies, tools and data-sets used throughout the thesis are detailed in Chapter 2 Data and Modelling . Chapter 3 Biogenic Isoprene emissions in Australia shows how models are misrepresenting isoprene emissions by a large margin in Australia, along with the description and implementation of a relatively simple method of improvement. Chapter 4 Stratospheric ozone intrusions provides an estimate of stratospheric ozone influx to the troposphere, along with potential classifications and seasonality.

I aim to recalculate satellite vertical columns of HCHO using updated model a priori information. HCHO from OMI on board the AURA satellite is examined and recalculated in Chapter 2, where influence of the air mass factor (AMF) is discussed in detail. This AMF was created using an older version of GEOS-Chem with out-of-date HCHO chemistry. Recalculation, binning, and analysis of the satellite HCHO vertical columns is performed using GEOS-Chem v10.01, outlined in Section 2.6.1. This is performed using my own partial AMF recalculation code, along with full recalculation code set up in collaboration with Prof. Palmer and Dr. Surl. Subsequent use of the data makes further use of the recalculation as a basic sensitivity analysis of top-down emissions estimations to a priori satellite information.

I aim to determine biogenic isoprene emissions in Australia using a top-down inversion of satellite HCHO, through an estimated yield from GEOS-Chem. In Chapter 3 I determine the linear relationship between total column HCHO and biogenic isoprene emissions over Australia using the global CTM GEOS-Chem. Applying this relationship to satellite based HCHO measurements creates the desired top-down isoprene emissions estimate. This process is described in Section 3.2, and requires intensive filtering of both satellite data (to exclude non-biogenic HCHO sources) and model yield (to minimise spatial smearing). The uncertainty and limitations of top-down estimates due to satellite and model uncertainty, along with temporal and horizontal resolution of available data is examined in Section 3.4. Furthermore, this top-down estimation is used to scale a new simulation of HCHO, and O_3 over Australia,

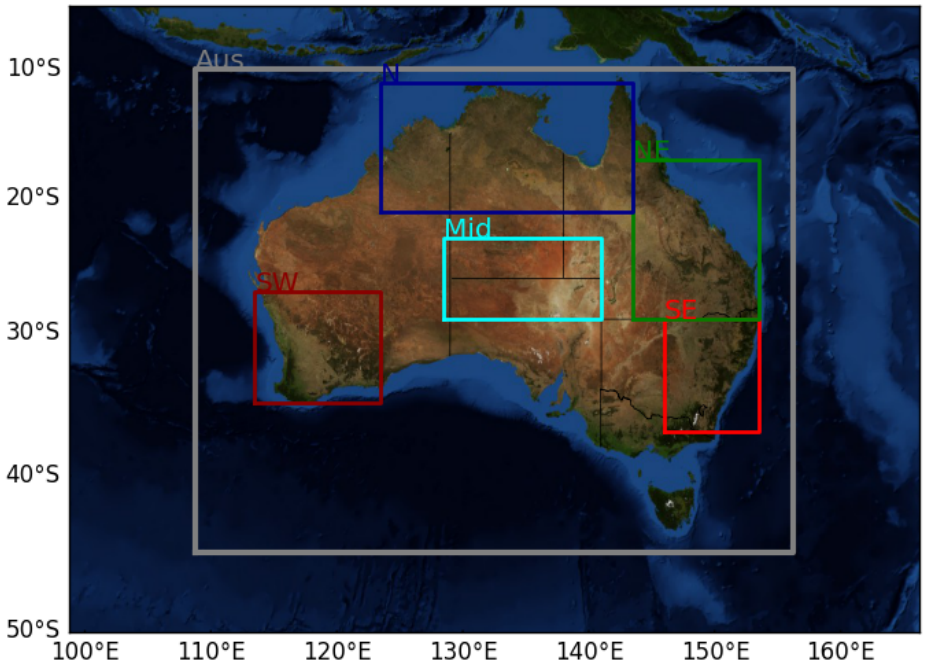

FIGURE 5.1: Sub-regions used in subsequent figures. Australia-wide averages will be black or grey, while results from within the coloured rectangles will match the colours shown here.

described in Section 3.2.8.

I aime to improve understanding of ozone transported to the troposphere from the stratosphere in Australia and the southern ocean. In Chapter 4 the seasonal cycle of STT events is characterised, and their contribution to the SH extra-tropical tropospheric ozone budget is quantified using GEOS-Chem to estimate ozone flux extrapolated from three measurement sites. Causal climatology and event seasonality are examined in Section 4.4. STT detection frequencies and modelled tropospheric ozone columns are used to estimate STT ozone flux near three sites in Section 4.6. Findings are compared against relevant literature, and the uncertainties involved in STT event detection and ozone flux estimation are studied.

In the following sections, summaries of results over Australia are often split into regions shown in Figure 5.1.

5.2 Isoprene emissions

Being a major driver of continental boundary ozone production, a new isoprene emissions estimate is created using OMI satellite measurements of HCHO. This estimate is and compared against the bottom-up estimate from GEOS-Chem (running MEGAN).

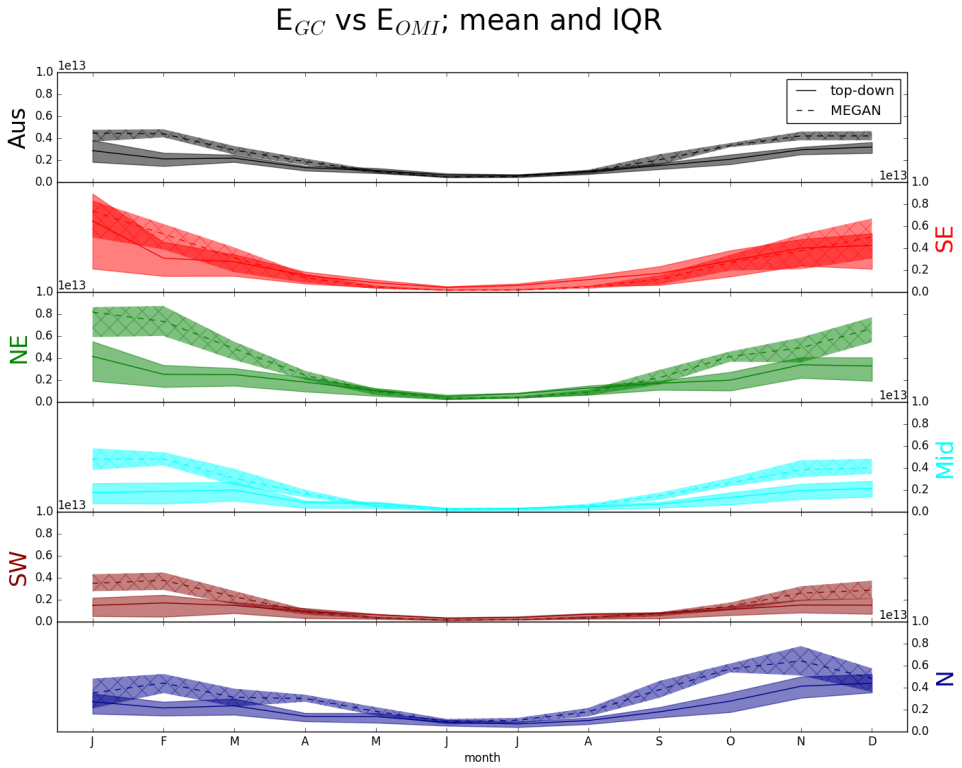

FIGURE 5.2: The multi-year monthly mean (lines) and IQR (shaded) of midday (13:00-14-00 LT) isoprene emissions estimates. Estimates come from MEGAN run by GEOS-Chem (E_{GC}), and the OMI top-down technique (E_{OMI}). The mean E_{GC} is shown by the dashed lines and hatched shaded areas show the IQR. E_{OMI} means are shown using the solid lines, with IQR shown by unhatched shaded areas. Colours denote the region over which the monthly average was taken, as shown in Figure 5.1.

Figure 5.2 shows these emissions compared regionally. Generally months outside of May to August show the a postiori lower than the a priori, except in the south eastern portion of Australia where only February is notably reduced. MEGAN estimates of isoprene emissions appear to be largely overestimated in Austral summer, suggesting poorly understood emission factors for Australian forests.

5.3 Ozone over Australia

Ozone production in the troposphere is a complex process involving various compounds. Tropospheric ozone is enhanced through VOC chemistry, stratospheric transport, and pollution. The first two of these processes are highly uncertain, with few studies performed on either topic within Australia. Of these ozone sources, VOC chemistry uncertainty is dominated by poor understanding of biogenic emissions (mainly) of isoprene. Emissions of isoprene are globally modelled at ~465-500 Tg C yr^{-1}(Guenther

TABLE 5.1: Isoprene emissions from MEGAN and top-down estimation in Tg a^{-1}, along with ozone tropospheric column amounts in O_3 $cm^{-2} \times 10^{17}$.

Metric	AUS	SEA	NEA	NA	SWA	MID
MEGAN	43(2)	blah				
Ozone	9.70	11.17	11.03	11.19	11.69	9.09
Top-Down	19(2)					
Ozone	9.64	11.11	10.99	11.12	11.63	9.02

Standard deviations shown in parenthesis.

et al. 2006; Messina et al. 2016). However these appear to be overestimated in Australia, as seen here with top-down estimates at $\sim$ 19 Tg C yr^{-1} compared to $\sim$ 43 Tg C yr^{-1} using GEOS-Chem (which implements MEGAN).

TODO: ozone concentrations using emissions from MEGAN/TOP-Down, also how these compare to estimated STT: Tropospheric ozone production is estimated using GEOS-Chem before and after scaling isoprene emissions based on top-down satellite data. Figure 5.3 shows Australian summer surface level ozone with and without scaling isoprene emissions to match the multi-year top-down estimation. Tropospheric column ozone reduces very minimally ($<$ 1%), however surface ozone drops $\sim$ 5%. This suggests that impacts from reduced isoprene emission do not propagate vertically. The results are tabulated for each of five regions, and all of Australia, and shown here in Table 5.1. Although isoprene emissions are halved on average over Australia, the reduction has only a small effect on ozone.

Ozone transported from the stratosphere also may have some affect on surface ozone levels, although this is hard to detect with the methods used in this thesis. We only compare the affect of the two disparate influences on the tropospheric ozone column. STT analysis over Melbourne suggests up to $\sim$ 10% of the tropospheric column is due to stratospheric influx, with the average increase caused by influx being $\sim$ 1 to 3.5%. We see that a relatively large drop in isoprene emissions causes $\sim$ 1% reduced ozone concentration over summer. Both of these sources become more important in summer, with more frequent STT and stronger isoprene emissions. However the impacts are separated by altitude with most STT occurring in the free troposphere, while isoprene emission impacts are largely seen at the surface.

TODO: how isoprene is linked to ozone in modelling Figure 5.4 shows surface level (up to $\sim$ 150 m altitude) ozone concentration over 2005 before and after scaling modelled isoprene emissions. Reducing isoprene emissions lowers surface ozone concentrations by TODO: XX to YY % in summer, and XX to YY % in winter. The direct correlation between reduced emissions and surface ozone (Figure TODO) is more or less clear monthly, and using multiple years enhances/reduces the relationship.

5.4 Outputs and future work

Analysis of STT along with an estimate of ozone flux from the stratosphere over a portion of Australia and the southern ocean is published in **Greenslade2017** Ozone

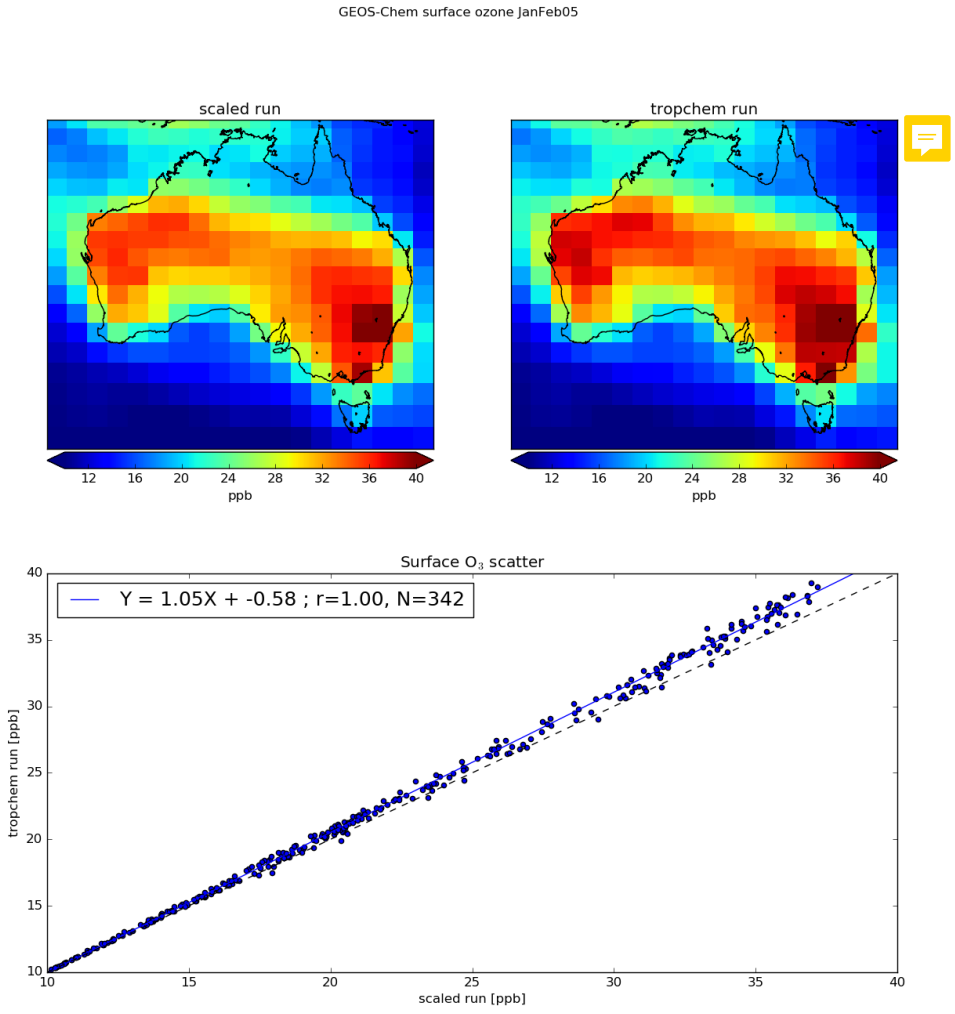

FIGURE 5.3: Ozone maps before (left) and after (right) scaling isoprene emissions in GEOS-Chem for summer. The bottom panel shows the linear regression between the runs along with a black dashed line representing the 1-1 ratio.

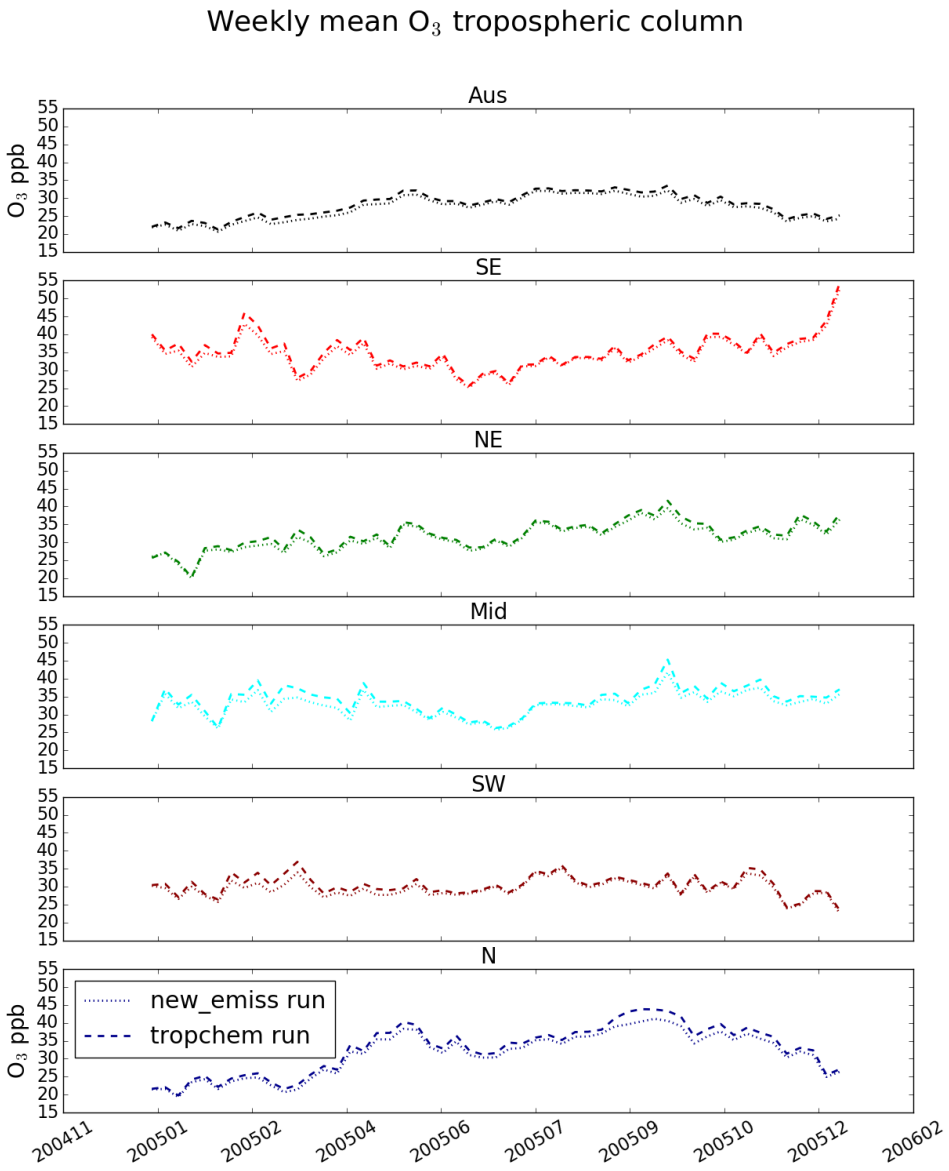

FIGURE 5.4: Surface ozone concentrations (ppb) per region over 2005.

production using GEOS-Chem with updated top-down isoprene emissions is examined in this thesis and may form part of a new publication in the near future with the aim of updating MEGAN isoprene emission factors in Australia.

One of the important parameters in Australia is the soil moisture activity factor(γ_{SM}), which can have large regional effects on the isoprene emissions (Sindelarova et al. 2014; Bauwens et al. 2016). Generally if soil moisture is too low, isoprene emissions stop (Pegoraro et al. 2004; Niinemets et al. 2010), however in many Australian regions the plants may be more adapted to lower moisture levels. (TODO: Find cites for this - talk from K Emerson at Stanley indicated this) GEOS-Chem runs MEGANv2.1, which has three possible states for isoprene emissions based on the soil moisture (θ):

$$\begin{aligned} \gamma_{\mathrm{SM}} &= 1 && \theta > \theta_1 \\ \gamma_{\mathrm{SM}} &= (\theta - \theta_w)/\Delta\theta_1 && \theta_w < \theta < \theta_1 \\ \gamma_{\mathrm{SM}} &= 0 && \theta < \theta_w \end{aligned}$$

where θ_w is the wilting point, and θ_1 determines when plants are near the wilting point. The wilting point is set by a land based database from Chen and Dudhia (2001), while θ_1 is set globally based on Pegoraro et al. (2004). These moisture states are disabled in GEOS-Chem V10.01. Improved isoprene emissions modelling requires this soil moisture problem to be handled. Simply enabling the parameter in its current form is not quite good enough for Australia, due to both the unknown soil moisture and the poorly understood plant responses in this country. TODO: cite the paper which could update MEGAN soil moisture parameterisation.

Using satellite data to improve isoprene emission estimates such as done in this thesis must be used to analyse model improvements, since fully independent measurements are lacking. Measurements of isoprene emissions in Australia would provide a valuable opportunity to verify improvements in emission estimates. Emissions measurement would be greatly valuable however they remain expensive and difficult, especially over the large sparse environment which makes up the Australia outback.